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Applicant:	Neumann *et al.*	Examiner:	Unassigned
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For:	Three-Dimensional Catalyst Gauzes Knitted in Two or More Layers		

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May 6, 2002

Commissioner for Patents
Washington, D.C. 20231

SUBMISSION OF TRANSLATION OF PRIORITY DOCUMENT AND STATEMENT OF ACCURACY OF TRANSLATION PURSUANT TO 37 C.F.R. § 1.78(a)(5)

Sir:

Pursuant to 37 C.F.R. § 1.78, Applicants hereby submit: (1) a translation of the priority document for the above referenced application, which was filed on February 15, 2001 as a provisional patent application and was assigned Provisional Patent Application Serial Number 60/268,718; and (2) a translator's declaration that attests to the accuracy of the translation.

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5/6/02
May 6, 2002

K Padilla
Kim Padilla

Applicants submit that no fee is necessary with this submission. However, if any fee is due, authorization is hereby given to charge Deposit Account No. 11-0171 for such sum.

If there are any questions or comments relating to this submission, the Examiner is respectfully invited to contact Applicants' attorney at the telephone number set forth below.

Respectfully submitted,

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TRANSLATOR'S DECLARATION

I, Janet Hope, BSc.(Hons.), MIL., MITI., translator to Messrs. Taylor & Meyer of 20 Kingsmead Road, London, SW2 3JD, Great Britain, verify that I know well both the German and the English language, that I have prepared the attached English translation of 17 pages of a German Patent application in the German language with the title:

Dreidimensionale, zwei- oder mehrlagig gestrickte Katalysatornetze für Gasreaktionen

identified by the code number 000777 EM at the upper left of each page and that the attached English translation of this document is a true and correct translation of the document attached thereto to the best of my knowledge and belief.

I further declare that all statements made of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements are made with the knowledge that wilful false statements and the like are punishable by fine or imprisonment, or both, under 18 USC 1001, and that such false statements may jeopardize the validity of this document.

By: ______________ Date: ______________

Three-dimensional catalyst gauzes for gas reactions knitted in two or more layers

Description:

The invention relates to three-dimensional catalyst gauzes for gas reactions knitted in two or more layers from noble metal wires, in which the meshes of the individual layers are joined to one another by pile threads and weft threads are inserted between the mesh layers.

Gas reactions catalysed by noble metals, such as the oxidation of ammonia with atmospheric oxygen in the production of nitric acid (Ostwald process) or the reaction of ammonia with methane in the presence of oxygen to give hydrocyanic acid (Andrussow process) have acquired considerable industrial importance for a long time, and indeed base chemicals for the chemical industry and for fertilizer production are provided by them on a large industrial scale.

Noble metal catalysts in the form of gas-permeable spatial structures on or in which the reaction proceeds are the core of these heterogeneously catalysed gas reactions. Gauzes in the form of woven fabrics or knitted fabrics of fine noble metal wires have found use here for a long time. The noble metal wires are made predominantly of platinum, rhodium or of alloys of these metals with other noble or base metals. Platinum-rhodium alloys with 4 to 12 wt.% rhodium and platinum-palladium-rhodium alloys with 4 to 12 wt.% palladium and rhodium are typical here. Palladium-nickel alloys with 2 to 15 wt.% nickel, palladium-copper alloys with 2 to 15 wt.% copper and palladium-nickel-copper alloys with 2 to 15 wt.% nickel and copper are furthermore employed.

The catalyst gauzes are conventionally arranged here in the reaction zone of a flow reactor in a plane perpendicular to the direction of flow of the gas mixture. Conical arrangements are also known. Several gauzes are usually expediently arranged in series and combined to form a so-called catalyst pack. Platinum-collecting gauzes, so-called getter gauzes, are conventionally arranged downstream in the catalyst pack, after the actual catalyst gauzes, these serving to recover platinum and rhodium discharged from the catalyst gauzes in the form of gaseous oxides convectively with the reaction gas stream. These getter gauzes are usually made of wires of palladium or palladium alloys.

Figure 1 shows a diagram of the reactor with the reaction proceeding therein by the example of catalytic oxidation of ammonia.

In the reaction zone (2) of the flow reactor (1), the catalyst pack (3), which comprises several catalyst gauzes (4) in series and downstream getter gauzes (5), is arranged in a plane perpendicular to the direction of flow. The ammonia/atmospheric oxygen mixture (with an ammonia content of 9 - 13 vol.%) (6) flows through the catalyst pack under atmospheric or increased pressure, ignition of the gas mixture taking place in the entry region and the combustion reaction to give nitrogen monoxide (NO) and water (7) involving the entire catalyst pack. The NO in the reaction gas mixture (7) flowing out subsequently reacts with the excess atmospheric oxygen to give NO_2 (8), which forms nitric acid with water in a downstream absorption (9), the product being fed, for example, to fertilizer production.

Knitted noble metal catalyst gauzes have a number of advantages over woven catalyst gauzes, and for this reason they are currently preferred in industrial use. On the one hand, knitted catalysts can be produced more economically, since shorter set-up times are achieved with the knitting technique than with the weaving technique. This causes in

particular a considerably reduced binding of noble metal in production. Using the flat bed knitting technique, the knitted gauzes are produced in individual production in shape and to dimensions, while woven gauzes have to be cut out from finished webs, expensive waste being produced. The knitting technique also offers the possibility of a high flexibility in respect of knitting pattern, wire thicknesses used and resulting weight per unit area. On the other hand, three-dimensionally knitted catalyst gauzes in particular prove to be more effective catalytically than gauzes knitted in one layer or even woven gauzes because of their more complex spatial structure.

This applies above all to the three-dimensional catalyst gauzes knitted in two or more layers which are described in EP 0 680 767 and in which the meshes of the individual layers are joined to one another by pile threads.

Nevertheless, such three-dimensionally knitted catalyst gauzes are in need of still further improvement in respect of catalytic activity, selectivity of the reaction catalysed, amount of noble metal employed, mechanical strength, service life and unavoidable loss of noble metal. In addition to these economic requirements, on the ecological side the requirement to reduce the N_2O emissions arising on the catalyst gauzes is of primary importance. The problem here is chiefly that for a complete conversion of ammonia, an adequate residence time of the reaction gas in the catalyst pack and a corresponding porosity of the catalyst pack is necessary. The complete conversion of ammonia in the Ostwald process is absolutely necessary, since ammonium nitrites and nitrates which are an explosion hazard can form if unreacted ammonia passes through the catalyst pack. The mechanical stability of the catalyst gauzes must furthermore be ensured in respect of the required service life. On the basis of these basic requirements of the catalyst gauze and catalyst pack, there

is a given minimum number of catalyst gauzes and a minimum wire thickness thereof, which predetermines the minimum amount of noble metal employed. However, the weight per unit area of the gauzes also cannot be reduced as desired, for example by reducing the wire thickness, since this would have an adverse effect on the mechanical strength and the service life of the gauzes. A reduction in the processed wire length would result in a widening of the mesh width in the catalyst gauzes which are conventional nowadays, as a consequence of which the proportion of unreacted ammonia which passes through in this gauze layer increases. A reduced reactivity of such gauzes furthermore leads to an increased formation of N_2O, especially in the start-up phase of the reactor.

The present invention was therefore based on the object of further increasing the catalytic activity and efficiency of noble metal catalyst gauzes for gas reactions such that it is possible to manage with a lower total amount of noble metal employed, for example by reducing the number of gauzes and/or the length of the wire processed in the catalyst gauze and/or the wire thickness thereof, without thereby having to accept disadvantages in respect of the yield and selectivity of the gas reaction, mechanical strength and service life of the gauzes and unavoidable loss of noble metal.

This object is achieved according to the invention by three-dimensional catalyst gauzes for gas reactions knitted in two or more layers from noble metal wires, in which the meshes of the individual layers are joined to one another by pile threads and which are characterized in that weft threads are inserted between the mesh layers.

The basic structure of the catalyst gauzes according to the invention corresponds to the three-dimensional catalyst gauzes knitted in two or more layers described in EP 0 680 767. In these the meshes of the individual layers

are joined to one another by pile threads. Up to ten pile threads per mesh can be present here, the pile threads being aligned at an angle of 0 to 50° to the direction of flow of the reaction gases (corresponding to 90 to 40° to the plane of the gauze). The pile threads typically have a length of 1 to 10 mm. Corresponding two-layer knitted fabrics have a thickness of 1.0 to 3.0 mm and a weight per unit area of 1000 to 3000 g/m^2.

According to the invention, weft threads are now additionally inserted between the mesh layers. The weft threads can be inserted between the mesh layers in several planes. The weft threads are preferably arranged approximately centrally between two mesh layers. The weft threads are typically arranged here unidirectionally in the planes. The weft threads are preferably arranged approximately parallel to one another and are aligned in their direction perpendicular to the direction of the meshes in the mesh layers. The weft threads are expediently inserted into the pile threads which join the mesh layers and are fixed by these. The weft threads can also be constructed as multiple wires.

The knitted catalyst gauzes according to the invention typically have a number of weft threads per mesh corresponding to their wire properties.

The weft threads are made of the same wire material as the mesh and pile threads, namely preferably of platinum-rhodium alloy with 4 to 12 wt.% rhodium and platinum-palladium-rhodium alloys with 4 to 12 wt.% palladium and rhodium. Typical such alloys are PtRh5, PtRh8 and PtRh10.

Wires which have a diameter of 0.05 to 0.120 mm and which have a tensile strength of 900 to 1050 N/mm^2 and an elongation limit of 0.5 to 3% are preferably employed for knitting the gauzes according to the invention. The expert is familiar with the production of wires from corresponding

noble metal alloys by linear cold forming. Such wires can be processed without auxiliaries on flat bed knitting machines in accordance with EP 0 504 723.

In the knitted catalyst gauzes according to the invention, the mesh threads, pile threads and weft threads can have thicknesses which differ from one another. Typically, independently of one another, the mesh threads have wire diameters of 0.06 to 0.092 mm, the pile threads have wire diameters of 0.06 to 0.092 mm and the weft threads have wire diameters of 0.06 to 0.092 mm.

In the knitted catalyst gauzes according to the invention, the mesh threads, pile threads and weft threads can be reduced in minimum wire thickness by up to 15%. The wire length processed in the mesh and pile threads can in each case be reduced here by up to 50%. Of the amount of noble metal saved as a result, at least 40% is inserted into the catalyst gauze in the form of weft threads. No disadvantages arise in respect of yield and selectivity of the gas reaction, mechanical strength and service life of the gauzes and unavoidable loss of noble metal.

The knitted catalyst gauzes according to the invention can be produced on commercially available industrial flat bed knitting machines (e. g. from Stoll, Reutlingen, type CSM 440 TC) by also running a weft thread guide between the mesh thread guide and the pile thread guide. In accordance with EP 0 504 723, the settings on the flat bed knitting machines are preferably between 3.63 and 1.81 mm in respect of gauge and between 2 and 6 mm for the mesh length.

Figure 2 shows a magnified diagram of a section from a knitted catalyst gauze according to the invention. In the graphical diagram, the pile and weft threads are represented with a larger wire thickness than the mesh thread for visual illustration of the structure of the gauze geometry. The figure shows a catalyst gauze of two

mesh layers (2), (3) joined to one another by pile threads (1), into which weft wires (4) arranged approximately parallel to one another are inserted as single wires approximately centrally between the mesh layers (2), (3). The weft wires (4) are fixed in the crossing points (5) of the pile threads (1) and form a further catalytically active plane there approximately centrally between the mesh layers (2), (3).

By introducing the weft wires, an additional dense noble metal wire plane is inserted into the three-dimensional spatial structure of the knitted fabric at the pile threads which cross over each other, as a result of which the rate of reaction in the catalyst gauze is increased. The weft wires are fixed by the pile threads which cross over each other, so that further stabilization of these wires by linking via the formation of meshes is unnecessary. Compared with a corresponding catalyst gauze in one layer, this involves a significantly lower amount of noble metal employed due to the plane formed by the weft wires.

It is found that the knitted catalyst gauzes according to the invention have a significantly higher catalytic activity than conventional three-dimensional catalyst gauzes knitted in two or more layers (corresponding to EP 0 680 767) into which no weft wires are inserted. Gas reactions can thus be operated either with a lower number of catalyst gauze layers in the catalyst pack and/or with gauzes made of noble metal wires of shorter processing length or smaller thickness, depending on whether they are conducted under atmospheric pressure or under pressure. This results in a significantly lower total amount of noble metal employed. The reduction in the amount of noble metal employed is between 15 and 30%.

The advantageous nature of the catalyst gauzes according to the invention also manifests itself in the ignition properties of the catalyst pack and during the critical

start-up phase of the reaction. As a result of the higher catalyst activity, the ignition temperature is lowered, typically by 20 to 30°C, and the operating temperature of the catalyst pack of 800 to 950°C is therefore reached considerably faster. The time required to achieve a stable reaction is typically reduced by 20 to 50%. The N_2O emission, in particular in the start-up phase of the reaction, is thus lowered by on average 15 to 30% and the product yield is increased accordingly.

Example 1:

A research reactor for oxidation of ammonia is operated under conditions typical for medium-pressure plants (pressure: 4.0 bar; operating temperature: 860°C; throughput of ammonia: 0.12 m^3/h) in each case with a catalyst pack, diameter 12 mm, of the following configuration:

(a) combination of (conventional, prior art):

3 catalyst gauzes knitted in one layer of PtRh8; wire thickness 0.076 mm; weight per unit area 600 g/m^2

1 catalyst gauze knitted in two layers of PtRh8; wire thicknesses: mesh thread 0.076 mm, pile thread 0.076 mm; gauze thickness 2.5 mm; weight per unit area 1800 g/m^2

(b) combination of (modified according to the invention):

3 catalyst gauzes knitted in one layer of PtRh8; wire thickness 0.076 mm; weight per unit area 600 g/m^2

1 catalyst gauze according to the invention knitted in two layers of PtRh8; wire thicknesses: mesh thread 0.076 mm, pile thread 0.076 mm, weft thread 0.076 mm; gauze thickness 2.5 mm; weight per unit area 1800 g/m^2

The ignition temperature of the catalyst pack modified according to the invention is 230°C and therefore 20 - 30°C below that of the conventional catalyst pack. In the start-up phase of the catalyst pack modified according to the invention, the N_2O emission is lowered by 20%. In both cases the operating temperatures are established almost immediately after the ignition. While a stationary operating state with constant product distribution is established after the operating temperature is reached with the catalyst gauze according to the invention, this is achieved only after 0.5 to 3.5 hours with the conventional catalyst pack.

Example 2:

An industrial reactor for oxidation of ammonia is operated under conditions typical for medium-pressure plants (pressure: 6.3 bar; operating temperature: 895°C; throughput of ammonia: 5121 m^3/h) with a catalyst pack, diameter 1700 mm, of the following configuration:

(a) combination of (conventional, prior art):

3 catalyst gauzes knitted in one layer of PtRh5; wire thickness 0.076 mm; weight per unit area 600 g/m^2

4 catalyst gauzes knitted in two layers of PtRh5; wire thickness 0.076 mm; weight per unit area 1800 g/m^2

Total weight of noble metal incorporated 20.5 kg.

(b) combination of (modified according to the invention):

2 catalyst gauzes knitted in one layer of PtRh5; wire thickness 0.076 mm; weight per unit area 600 g/m^2

3 catalyst gauze [sic] knitted in two layers of PtRh5; wire thickness 0.076 mm; weight per unit area 1800 g/m^2

1 catalyst gauze according to the invention knitted in two layers of PtRh5; wire thicknesses: mesh thread 0.060 mm, pile thread 0.060 mm, weft thread 0.060 mm; gauze thickness 2.55 mm; weight per unit area 1600 g/m^2

Total weight of noble metal incorporated 16.5 kg.

The catalyst pack according to the invention comprises a total of 6 catalyst gauzes, of which 1 is a catalyst gauze according to the invention knitted in two layers with weft threads. The conventional catalyst pack of comparable efficiency comprises 7 gauzes, of which 3 are catalyst gauzes knitted in one layer and 4 are catalyst gauzes knitted in two layers (corresponding to EP 0 680 767). The catalyst gauze according to the invention results in a reduction in the total amount of noble metal employed by 20% from 20.5 kg to 16.5 kg.

The reduction in the amount of noble metal employed by the catalyst gauze according to the invention knitted in two layers is composed as follows:

1 catalyst gauze knitted in one layer with a wire thickness of 0.076 mm and a weight per unit area of 600 g/m^2 and 1 catalyst gauze knitted conventionally in two layers with a wire thickness of 0.076 mm and a weight per unit area of 1800 g/m^2 were replaced by 1 catalyst gauze according to the invention knitted in two layers with a wire thickness of 0.060 mm and a weight per unit area of 1600 g/m^2. The reduction in weight is 1.816 kg (33%), where of this [sic] 1.362 kg (75%) of the weight reduction is to be attributed to the reduction in the number of gauzes in the catalyst pack and 0.454 kg (25%) is to be attributed to the

reduction in the wire thickness in the catalyst gauze according to the invention knitted in two layers.

The further saving of 2.184 kg for the total catalyst pack was due to a reduction in the wire thickness and the weight per unit area of 2 of the 3 conventional two-layer catalyst gauzes employed.

The ignition temperature of the catalyst pack cannot be measured in this plant. The operating temperature is reached after approx. 2 minutes. This is about 60% of the start-up time required with conventional catalyst packs. The ammonia conversion after the operating temperature has been reached is complete in both cases.

After an operating period of 4 weeks, a stable yield 1% higher is achieved with the catalyst gauzes according to the invention.

Three-dimensional catalyst gauzes for gas reactions knitted in two or more layers

Patent claims:

1. Three-dimensional catalyst gauzes for gas reactions knitted in two or more layers from noble metal wires, in which the meshes of the individual layers are joined to one another by pile threads,
characterized in that
weft threads are inserted between the mesh layers.

2. Catalyst gauzes according to claim 1,
characterized in that
the weft threads are inserted between the mesh layers in several planes.

3. Catalyst gauzes according to claim 1 or 2,
characterized in that
the weft threads are arranged approximately centrally between two mesh layers.

4. Catalyst gauzes according to claims 1 to 3,
characterized in that
the weft threads are arranged approximately parallel to one another and are aligned in their direction perpendicular to the direction of the meshes in the mesh layers.

5. Catalyst gauzes according to claims 1 to 4,
characterized in that
the weft threads are inserted into the pile threads which join the mesh layers and are fixed by these.

6. Catalyst gauzes according to claims 1 to 5,
characterized in that,
independently of one another, the mesh threads have wire

diameters of 0.06 to 0.092 mm, the pile threads have wire diameters of 0.06 to 0.092 mm and the weft threads have wire diameters of 0.06 to 0.092 mm.

7. Catalyst gauzes according to claims 1 to 6, characterized in that up to ten pile threads per mesh are present and the pile threads are aligned at an angle of 0 to 50° to the direction of flow of the reaction gases (corresponding to 90 to 40° to the plane of the gauze).

8. Catalyst gauzes according to claims 1 to 7, characterized in that in the case of two-layer knitted fabrics they have a thickness of 1.0 to 3.0 mm and a weight per unit area of 1000 to 3000 g/m^2.

9. Catalyst gauzes according to claims 1 to 8, characterized in that the mesh threads, the pile threads and the weft threads are made of platinum-rhodium alloy with 4 to 12 wt.% rhodium or platinum-palladium-rhodium alloys with 4 to 12 wt.% palladium and rhodium.

10. Process for the production of three-dimensional catalyst gauzes according to claims 1 to 9, knitted in two or more layers from noble metal wires, on flat bed knitting machines, characterized in that in the knitting operation a weft thread guide is also run between the mesh thread guide and the pile thread guide.

11. Process according to claim 10, characterized in that wires which have a diameter of 0.05 to 0.120 mm and which have a tensile strength of 900 to 1050 N/mm^2 and

an elongation limit of 0.5 to 3% are employed for knitting the gauzes.

12. Process according to claim 11, characterized in that the settings on the flat bed knitting machines are between 3.63 and 1.81 mm in respect of gauge and between 2 and 6 mm for the mesh length.

13. Use of the catalyst gauzes according to claims 1 to 9 for carrying out heterogeneously catalysed gas reactions.

14. Use according to claim 11 in the oxidation of ammonia with atmospheric oxygen in the production of nitric acid (Ostwald process).

15. Use according to claim 11 in the reaction of ammonia with methane in the presence of oxygen to give hydrocyanic acid (Andrussow process).

Three-dimensional catalyst gauzes for gas reactions knitted in two or more layers

Abstract:

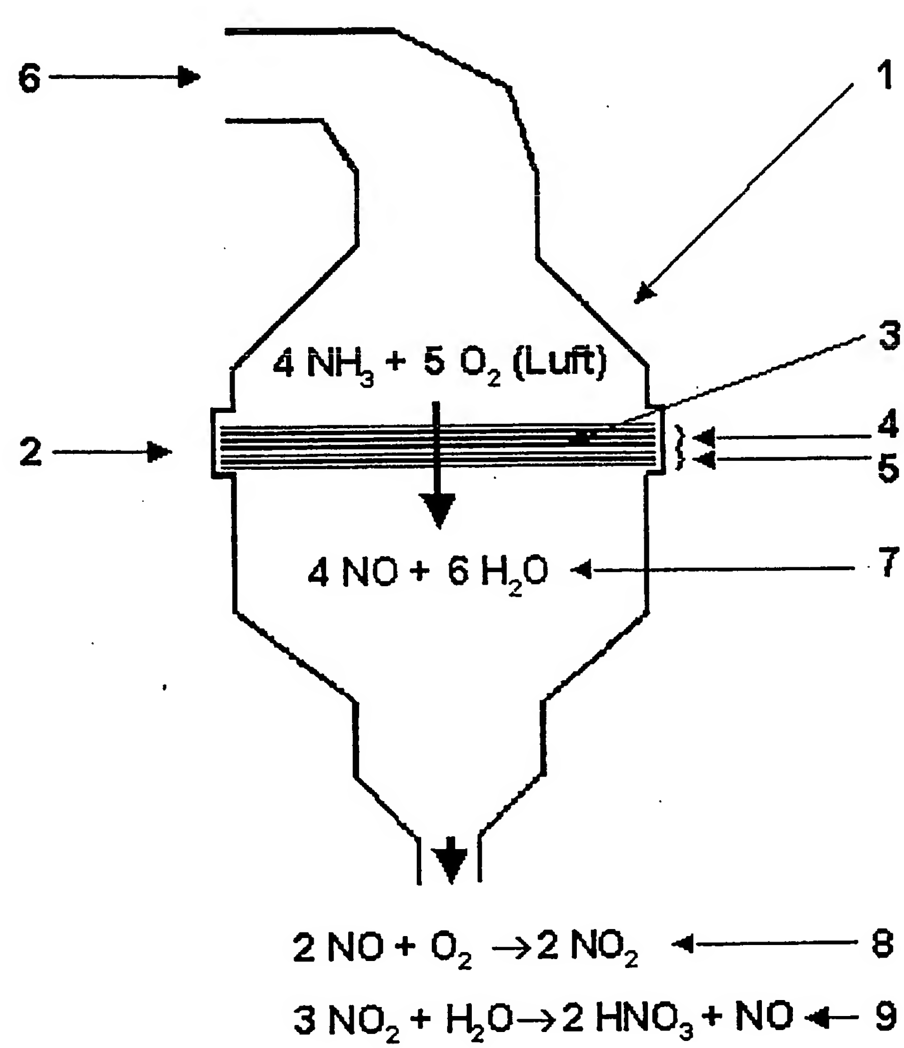
The invention relates to three-dimensional catalyst gauzes for gas reactions knitted in two or more layers from noble metal wires, in which the meshes of the individual layers are joined to one another by pile threads and which are characterized in that weft threads are inserted between the mesh layers.

These noble metal catalyst gauzes have an increased catalytic activity and efficiency in gas reactions, so that it is possible to manage with a lower total amount of noble metal employed, for example by reducing the number of gauzes and/or the length of the wire processed in the catalyst gauze and/or the wire thickness, without thereby having to accept disadvantages in respect of the yield and selectivity of the gas reaction, mechanical strength and service life of the gauzes and unavoidable loss of noble metal.

(Fig. 2)

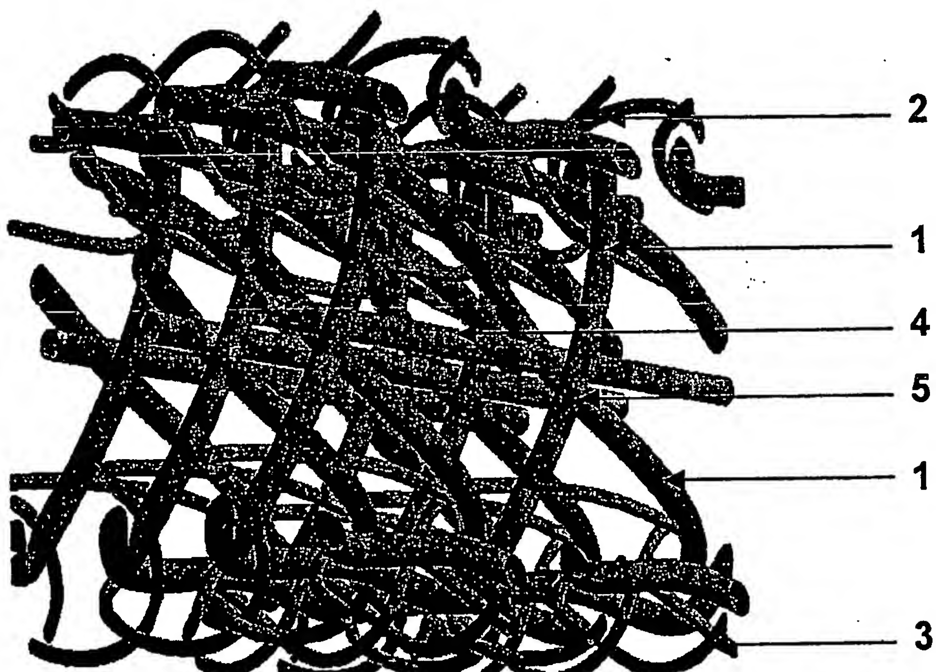
Three-dimensional catalyst gauzes for gas reactions knitted in two or more layers

Fig. 1



Three-dimensional catalyst gauzes for gas reactions knitted in two or more layers

Fig. 2



Translator's notes

The following errors have been found in the German:

Page 10, line 9 - "Katalysatornetz" (catalyst gauze) should read "Katalysatornetze" (catalyst gauzes).

Page 11, line 7 - "davon" appears to be superfluous.